

# Theoretical Study of the Effect of Hydroxy Subgroup on the Electronic and Spectroscopic Properties of Azulene Molecule: As a Nano Structure DFT Calculations

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## Abstract

Theoretical studies on Nono hydroxyl azulene molecules group in different positions were performed using DFT. Based on B3LYP with 6-31(d,p) basis set was used to investigate the effect of different position of hydroxyl (electron-withdrawing groups) on the electronic and structural properties of Nano hydroxyl azulene molecules. The optimized orientation and optimized parameters show that these structures have nano dimensions compared with the original molecule. The optimized structure, total energies, electronic states, energy gaps, ionization potentials, electron affinities, chemical potential, global hardness, softness, global electrophilicity, dipole moment and dipole polarizability were calculated. The harmonic vibration frequencies calculated and analyzed. The results showed a decrease in gap energies and improve the electronic properties.

**Keywords:** B3LYP/DFT, Ionization potential, electron affinity, energy gap, and IR spectrum

## Introduction

The organic semiconductor materials, such as, conjugated organic molecules have been widely used as active materials for optoelectronic devices such as light emitting diodes [3-5], field effect transistor [6-9], and photovoltaic and solar cells [10,11]. Aromatic compounds are important in industry and play key roles in the biochemistry of all living things [1], nitro-substituted on aromatic molecules are members of a class of environmental contaminants found in airborne particulate matter, fossil fuel combustion products, coal fly ash, cigarette smoke, and vehicular emissions, formed by reactions of aromatic molecules with nitrogen oxide [2]. These materials have advantages of easy fabrication, mechanical flexibility and low cost. There are many organic materials show useful field effect transistor performance, which can be characterized by their carrier mobility and on/off current ratios [13, 14]. The organic thin film transistors are one type of the so called organic devices, in which they fabricated by using the organic semiconductors [12].

Various studies on cyclic oligomers have been reported both experimentally and theoretically [15, 16], in [17] the substituent effects of oligomers such as oligothiophene, oligopyrrole and oligofuran are discussed in terms of reorganization energy. So, [18] studied the geometric and electronic properties for cyanothiophene oligomers as a prototype of an organic conducting polymer using ab initio and DFT [15], they showed that the cyngroup generally reduced the band gap with variation of the substitution position.

Azulene molecule and its derivatives have been studied in this work. Therefore the purpose of this work is to examine the hydroxy group substituent effect on azulene molecule varying the number of the substituent in the molecule.

## Theory and computational details

The structures of the molecules under study in this work are shown in figure 1. All the computational studies were carried out using the density functional theory (DFT) methods implemented in the Gaussian 09 suite of programs [3]. The molecular properties of the compounds had been computed by DFT using the standard 6-31G(d,p) basis set. Lee-Yang - Parr correlation functional [9] is used together with Becke's three parameters [3] exchange functional B3LYP. Conformational analysis of the molecules had been performed to have an idea about the lowest energy structures of the species.

The geometrical structure was performed at the B3LYP density functional theory with the same basis set [2, 4]. Harmonic vibration frequencies were computed at the same level of theory. The hybrid functional B3LYP has shown to be highly successful for calculation the electronic properties such as ionization potentials, electronic states and energy gaps [2-7]. The electronic energy as  $E = E_T + E_V + E_J + E_{XC}$ , where  $E_T$ ,  $E_V$ , and  $E_J$  are the electronic kinetic energy, the electron nuclear attraction and the electron-electron repulsion terms respectively. The electron correlation is taken into account in DFT due to the exchange correlation term  $E_{XC}$ , which includes the exchange energy arising from the anti-symmetry of the quantum mechanical wave function and the dynamic correlation in the motion of individual electrons; it makes DFT dominant over the conventional HF procedure [8].

The geometry optimized structures are obtained by restricted closed-shell formalism and without any symmetry restriction, and vibration analysis for each structure does not yield any imaginary frequencies, which indicates that the structure of each molecule corresponds to at least a local minimum on the potential energy

surface[4]. In this investigation, the more relevant electronic potential (IP), electron affinities (EA), chemical potential ( $\mu$ ) (the negative of electronegativity ionization potential is calculated as the energy difference between the energy of the molecule derived from electron-transfer and the respective neutral molecule;  $IP_v = E_{\text{cation}} - E_n$ ). The EA was computed as the energy difference between the neutral molecule and the anion molecule:  $EA = E_n + E_{\text{anion}}$  [3]. The HOMO and LUMO energy was also used to estimate the IP and EA in the framework of Koopmans' theorem:  $IP = -\epsilon_{\text{HOMO}}$  and  $EA = -\epsilon_{\text{LUMO}}$  [11].

In the density functional theory (DFT), one of the global quantities is chemical potential ( $\mu$ ), which is measures the escaping tendency of an electronic cloud, and equals the slope of the Energy versus N (number of electrons) curve at external potential  $v(r)$  [2]:

$$\mu = \left[ \frac{\partial E}{\partial N} \right]_{v(r)} \quad (1)$$

The finite difference approximation to Chemical Potential gives,

$$\mu \approx -\chi = -(IP + EA)/2 \quad (2)$$

The theoretical definition of chemical hardness has been provided by the density functional theory as the second derivative of electronic energy with respect to the number of electrons N, for a constant external potential  $v(r)$  [12]:

$$\eta = \frac{1}{2} \left[ \frac{\partial^2 E}{\partial N^2} \right]_{v(r)} = \frac{1}{2} \left[ \frac{\partial \mu}{\partial N} \right]_{v(r)} \quad (3)$$

The finite difference approximation to Chemical hardness gives,

$$\eta = (IP - EA)/2 \quad (4)$$

The softness is given as [31]:

$$S = \frac{1}{2\eta} = \left( \frac{\partial^2 N}{\partial E^2} \right)_{v(r)} = \left( \frac{\partial N}{\partial \mu} \right)_{v(r)} \quad (5)$$

The electrophilicity index is a measure of energy lowering due to maximal electron flow between donor and acceptor. Electrophilicity index ( $\omega$ ) is defined as [6],

$$\omega = \frac{\mu^2}{2\eta} \quad (6)$$

## Results and discussion

### I. Molecular geometry

The optimized structure parameters of molecules calculated by DFT-B3LYP levels with the 6-31G(d, p) basis set are listed in the table 1, in accordance with the atom numbering scheme given for molecule No.2 in figure 1. Table 1 shows the calculated bond lengths and angles for the studied molecules. From the theoretical values, we can find that most of the optimized bond angles are slightly larger than the experimental values for the C-C, C=C and C-H bonds in aromatics

### II. Energies.

Table (2) shows the values of the total energy and electronic states for the analyzed structures and the energy gap ( $\epsilon_{\text{LUMO}} - \epsilon_{\text{HOMO}}$ ) of the studied molecules. The total energy for all studied molecules as a linear function of OH side group number adding to the molecule. The final total energy of the product is the collection of total energy of all small molecules which build the product molecule, that means:

$$E_{\text{tot}} = E(\text{azulene}) + n E(\text{hydroxyl sub group}) \quad (9)$$

Where  $n$  is the number of OH sub group.

It is clear that from Table 2, the total energy is independent on the position of the hydroxy radical in the ring, and it is depends on the number of subsistent in the molecule. The substitution of hydroxy groups (electron withdrawing) causes few decreasing the HOMO and LUMO energy [7], and energy gap decreased. Therefore, the presence of substituent decreases the energy gaps improves the conductivities and also enhances the solubilities of these molecules.

The LUMO-HOMO energy gaps of tetra-hydroxy azulene molecule is less than that of the original molecule and the others, with decreasing energy gap, electrons can be easily excited from the ground state [35,36]. This effect of the side group was the largest in molecule **No.5**, it has energy gap of (3.148 eV). The energy gap of azulene (3.324 eV). The table 2 shows also the symmetry of studied molecules, the molecule **1** is planar with inversion center and have  $C_{2v}$  symmetry (high symmetry), and have lower electronegativity, while molecule **No.5** is planer and have  $C_s$  symmetry (low symmetry), and have higher electronegativity.

### III. Some important variables.

The used method B3LYP functional has a high efficient to calculate the electronic properties for the organic studied molecules, such as ionization potentials(IP), electron affinities(EA), electronegativity ( $\chi$ ), absolute hardness( $\eta$ ), absolute softness (S), electrophilicity index ( $\omega$ ). The properties that are displayed in Table 3 for each variable are computed by Koopmans' theorem, it is based on the differences between the HOMO and the LUMO energies of the neutral molecule and is known as orbital-vertical theory.

Table 3 clearly reveal that these hydroxyl azulene compounds have a tendency to capture electrons instead of donating them. The ionization potential for the mono-hydroxy azulene molecule is greater than that for the other molecules, but the origin azulene molecule has the largest value of ionization potential, this indicates that this molecule needs high energy to become cation comparing with the others. The strength of an acceptor molecule is measured by its electron affinity (EA) which the energy released when adding one electron to LUMO. An acceptor must have a high EA, adding the radical to the ring leads to decreasing the ability of the electron affinity for the molecule, EA for molecule **No.3** is the largest between the new structures, as we see in Table 3.

Few interesting observations have been made from the results that are shown in Table 3 obtained through the orbital-vertical method. The electron affinities (EA) computed from the energy of the lowest unoccupied molecular orbital (LUMO) are higher for all study molecules than that of the energy-vertical method. The ionization potential (IP) that results from the highest occupied molecular orbital is smaller for all study molecules than that of the energy-vertical method. From the previous investigations. This could be the reason for the low hardness values obtained from the orbital-vertical method. Koopman's theorem neglects the relaxation effect by using the frozen-orbital approximation. However, the Koopmans' theorem is a crude but useful and fast approach [9]. The behavior of electronegativity, softness and electrophilicity index for the studied molecules shows the magnitude smaller than these for the original ring, adding the radicals give the molecule more hard.

The molecules dipole moment represents a generalized measure of bond properties and charge densities in a molecule [10]. Molecule with electron acceptor group due to better charge distribution and increasing distance have higher dipole moment[32], from Table 4 molecule **No.6** has higher dipole moment (5.194 Debye) .

The results of the calculated polarizability for the studied molecules in table 4 showed that all substitution groups leads to increase the average polarizability and cause more reactive than the original molecules (**1**). The molecules **1** and **2** have average dipole polarizability equal 157.115 and 159.571a.u, they have the highest polarizability and have highest reactivity. This due to the ring delocalizing  $\pi$ -electron resonance from the phenyl groups [8].

## VI. IR Spectra

The IR spectra of (**1- 6**) molecules are provided in figure (2). The harmonic vibrational frequencies calculated for study molecules at B3LYP level using the 6-31(d, p) basis set. The (C – H) stretching vibrations of aromatic molecules in the region (2900 – 3250)  $\text{cm}^{-1}$  which is characteristic region for ready identification of (C – H) stretching vibrations and particularly the region (3250 – 3100)  $\text{cm}^{-1}$  for asymmetric stretching and (3100 – 2900)  $\text{cm}^{-1}$  for symmetric stretching modes of vibration [10].

The present work gives the frequency values at 3170, 3180, 3139.45 and 3206  $\text{cm}^{-1}$  for azulene which are consistent with experimental results of 3012, 3065, 3106 and 3136  $\text{cm}^{-1}$ . the aromatic molecules frequency has both in-plane (1100 – 1700)  $\text{cm}^{-1}$  and out-of-plane (below 1000  $\text{cm}^{-1}$ ) (C – H) bending vibrations, the in-plane vibration for azulene calculated at 894.6  $\text{cm}^{-1}$  and 962.48  $\text{cm}^{-1}$  which a good agreement with experimental results 878.3 and 954.9  $\text{cm}^{-1}$ [15], and the out-of-plane (C – H) deformation vibrations calculated at 617.407  $\text{cm}^{-1}$  and 758.48  $\text{cm}^{-1}$  is agreement with experimentally predicted in 603  $\text{cm}^{-1}$  and 726  $\text{cm}^{-1}$ [18].

It is clear from fig. (2) that the IR spectra for hydroxyl azulene molecules characters from that of azulene by multiply the vibration mode due to existing of (H – O) , (C – O) bonds, (C = C) stretching and (C – H) stretching. For (2 – 6) molecules the stretching of (C – O) bond has been observed at (1305 – 1410)  $\text{cm}^{-1}$ , the (C – H) stretching stay in the region (3100 – 3265)  $\text{cm}^{-1}$ , the stretching of (C – O) bond has been observed at (1300 – 1410)  $\text{cm}^{-1}$  and the stretching of (C = C) bond at the range (1400 – 1500)  $\text{cm}^{-1}$ , while the bending of (H – O) bond appeared at (1550 – 1685)  $\text{cm}^{-1}$ . The stretching (C – O) bond for molecules 5 are calculated at 1644.06  $\text{cm}^{-1}$  while the experimental value at 1743  $\text{cm}^{-1}$  [36]. The  $\pi$  electron density on the aromatic ring was delocalized in the presence of the -OH, which reduced the strength of (C = C) bond and depending on the position of nitro group. The torsion vibrations appear at very low frequency for (2 – 6) molecules at below 60  $\text{cm}^{-1}$ .

#### 4. Conclusions:

We have use DFT in this study to compute geometry optimization and electronic properties of azulene and hydroxyl azulene molecules group by using B3LYP function. The calculated electronic properties such as ionization potential, electron affinity, electronegativity, hardness, softness and electrophilic index by using orbital-vertical method, the important conclusions are:

- 1- Geometry optimization for molecule 1 has been found in a good agreement with experimental data, while for other studied molecules (2 – 6) it has not been found a reference data.
- 2- The total energies for hydroxyl azulene molecules group found not dependent on the position of the hydroxy radical in the ring and substitution hydroxyl group causes decreasing energy and more stability.
- 3- The presence of the substituent decreases the energy gap of the molecules under study, this is one of the important properties obtained in this work, and a small energy gap means small excitation energies of manifold of the excited states.
- 4- The electronic properties (IP, EA,  $\chi$ ,  $\eta$ , S,  $\omega$ ) was calculated by using orbital-vertical method, thus Koopman's theorem is useful method, but has large time for treatments.
- 5- The results showed that all substitution groups leads to increase the average polarizability and dipole moment and cause to more reactive than original molecules.
- 6- In IR spectra calculation shows a good agreement with experimental data for the original ring (molecules 1), adding the hydroxyl groups leads to increasing the vibration mode, and highest stretching vibration wave numbers and its gave suitable positions for OH with carbon atoms in phenyl ring.
- 7- Molecule 6 is the best option for n-type organic semiconductors because of its better LUMO – HOMO ratio and other electronic properties.

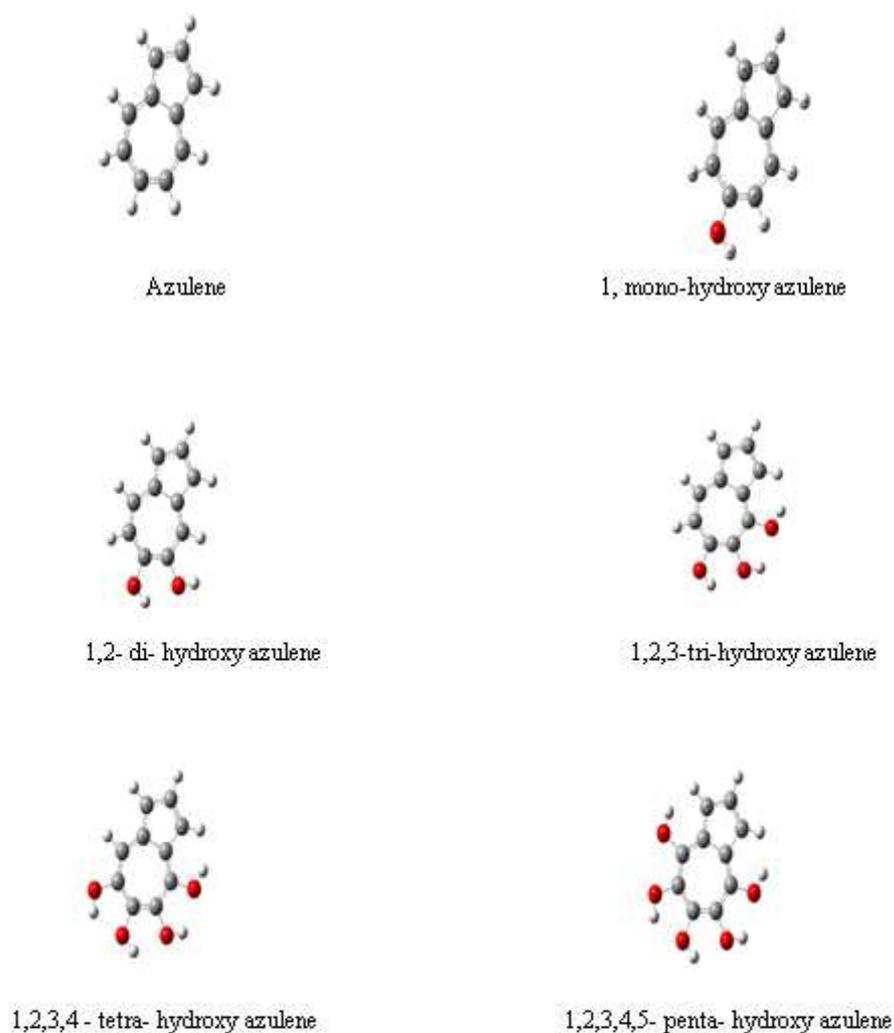
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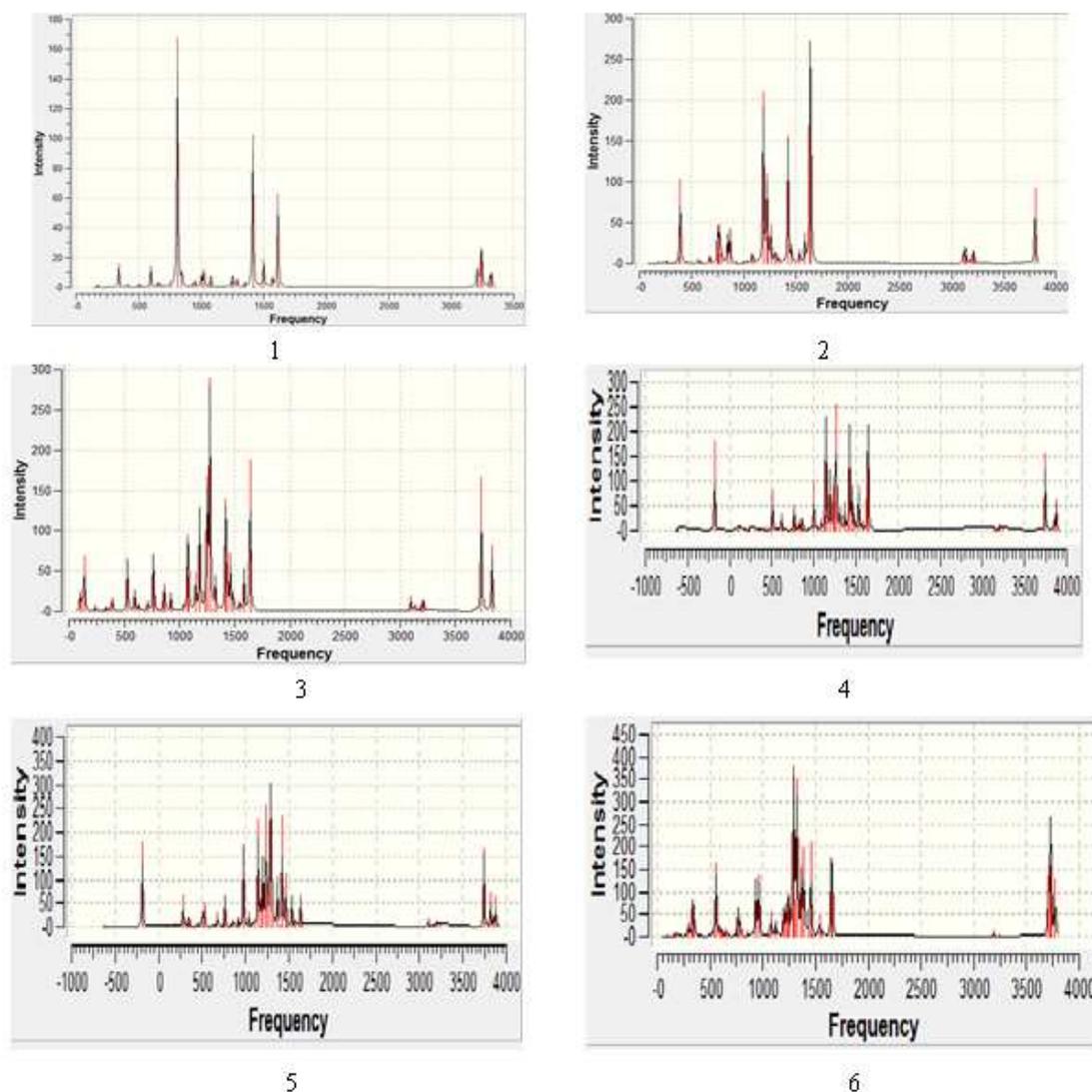
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**Figure 1.** Structures of azulene and its derivatives



**Figure 2:** The IR spectra of molecules under study, Epsilon  $\equiv$  Intensity (Km/mol).

**Table1: Optimized geometrical parameters of molecules, bond length R (Å) and angles (°).**

Molecules	bond length	Value	Angles	Value
1	R(C <sub>1</sub> -C <sub>2</sub> )	1.400	A(C <sub>2</sub> -C <sub>1</sub> -C <sub>3</sub> )	128.660
	R(C <sub>5</sub> -C <sub>13</sub> )	1.413	A(C <sub>1</sub> -C <sub>4</sub> -C <sub>6</sub> )	128.669
	R(C <sub>2</sub> =C <sub>3</sub> )	1.401	A(C <sub>5</sub> -C <sub>3</sub> -C <sub>13</sub> )	128.840
	R(C <sub>1</sub> -H <sub>8</sub> )	1.085	A(H <sub>8</sub> -C <sub>2</sub> -C <sub>3</sub> )	114.983
2	R(C <sub>1</sub> -C <sub>6</sub> )	1.430	A(C <sub>2</sub> -C <sub>3</sub> -C <sub>6</sub> )	120.396
	R(C <sub>2</sub> -C <sub>3</sub> )	1.427	A(C <sub>2</sub> -C <sub>1</sub> -N <sub>23</sub> )	117.373
	R(C <sub>1</sub> -N <sub>23</sub> )	1.47	A(C <sub>1</sub> -C <sub>2</sub> -H <sub>17</sub> )	118.767
	R(C <sub>2</sub> -H <sub>17</sub> )	1.084	A(C <sub>2</sub> -O <sub>8</sub> -H <sub>8</sub> )	125.788
	R(C <sub>1</sub> -C <sub>2</sub> )	1.423	A(C <sub>2</sub> -C <sub>1</sub> -C <sub>6</sub> )	122.761
3	R(C <sub>1</sub> -C <sub>6</sub> )	1.422	A(C <sub>2</sub> -C <sub>3</sub> -N <sub>23</sub> )	118.712
	R(C <sub>1</sub> -C <sub>3</sub> )	1.425	A(C <sub>1</sub> -C <sub>2</sub> -H <sub>17</sub> )	119.733
	R(C <sub>2</sub> -O <sub>8</sub> )	1.472	A(C <sub>14</sub> -C <sub>13</sub> -N <sub>21</sub> )	118.912
	R(C <sub>2</sub> -H <sub>17</sub> )	1.083	A(C <sub>2</sub> -O <sub>8</sub> -H <sub>8</sub> )	124.747
	R(C <sub>1</sub> -C <sub>2</sub> )	1.374	A(C <sub>2</sub> -C <sub>1</sub> -C <sub>6</sub> )	120.735
4	R(C <sub>1</sub> -C <sub>6</sub> )	1.415	A(C <sub>1</sub> -C <sub>2</sub> -N <sub>23</sub> )	115.521
	R(C <sub>2</sub> -C <sub>3</sub> )	1.442	A(C <sub>2</sub> -C <sub>1</sub> -H <sub>17</sub> )	118.488
	R(C <sub>2</sub> -O <sub>8</sub> )	1.473	A(C <sub>14</sub> -C <sub>15</sub> -N <sub>24</sub> )	115.524
	R(C <sub>1</sub> -H <sub>17</sub> )	1.082	A(C <sub>2</sub> -O <sub>8</sub> -H <sub>9</sub> )	123.679
	R(C <sub>1</sub> -C <sub>2</sub> )	1.368	A(C <sub>2</sub> -C <sub>1</sub> -C <sub>6</sub> )	120.669
5	R(C <sub>1</sub> -C <sub>6</sub> )	1.421	A(C <sub>3</sub> -C <sub>7</sub> -N <sub>23</sub> )	117.889
	R(C <sub>2</sub> -C <sub>3</sub> )	1.431	A(C <sub>2</sub> -C <sub>1</sub> -H <sub>16</sub> )	119.684
	R(C <sub>7</sub> -O <sub>9</sub> )	1.477	A(C <sub>8</sub> -O <sub>4</sub> -H <sub>9</sub> )	117.483
	R(C <sub>2</sub> -H <sub>17</sub> )	1.082	A(C <sub>2</sub> -O <sub>4</sub> -H <sub>12</sub> )	125.043
	R(C <sub>1</sub> -C <sub>2</sub> )	1.371	A(C <sub>2</sub> -C <sub>1</sub> -C <sub>6</sub> )	119.877
6	R(C <sub>1</sub> -C <sub>6</sub> )	1.409	A(C <sub>1</sub> -C <sub>2</sub> -N <sub>23</sub> )	115.105
	R(C <sub>1</sub> -C <sub>3</sub> )	1.438	A(C <sub>2</sub> -C <sub>1</sub> -H <sub>17</sub> )	119.437
	R(C <sub>2</sub> -O <sub>8</sub> )	1.478	A(C <sub>2</sub> -O <sub>23</sub> -H <sub>7</sub> )	118.449
	R(C <sub>2</sub> -H <sub>17</sub> )	1.082	A(C <sub>6</sub> -O <sub>23</sub> -H <sub>7</sub> )	124.452
	R(C <sub>1</sub> -C <sub>2</sub> )	1.371	A(C <sub>2</sub> -C <sub>1</sub> -C <sub>6</sub> )	119.877

**Table2: Total energy, electronic states and energy gap for molecules**

Structure	Energy(a.u)	Symmetry	Electronic States(eV)		Energy Gaps (eV)
			HOMO	LUMO	
1	-383.7826	C <sub>2v</sub>	-5.594	-2.270	3.324
2	-461.1120	C <sub>s</sub>	-5.492	-1.993	3.499
3	-536.3485	C <sub>s</sub>	-5.295	-2.054	3.241
4	-611.5643	C <sub>s</sub>	-5.265	-1.831	3.434
5	-686.7933	C <sub>s</sub>	-5.053	-1.904	3.148
6	-762.0060	C <sub>1</sub>	-5.155	-1.659	3.495

**Table3: The electronic properties for molecules.**

Molecules	IP(eV)	EA(eV)	X (eV)	H(eV)	S(eV) <sup>-1</sup>	W(eV)
1	5.594	2.270	3.932	1.662	0.300	4.6512
2	5.492	1.993	3.743	1.749	0.285	4.004
3	5.295	2.054	3.674	1.620	0.308	4.164
4	5.265	1.831	3.548	1.717	0.291	3.665
5	5.053	1.904	3.479	1.574	0.317	3.844
6	5.155	1.659	3.407	1.747	0.286	3.321

**Table 4:**calculated dipole moment  $\mu$ (debye), components of  $\alpha_i$  ( $i = xx,yy,zz$ ) and average of thedipole polarizability  $\langle \alpha \rangle$

molecules	$\mu$	$\alpha_{xx}$	$\alpha_{yy}$	$\alpha_{zz}$	$\langle \alpha \rangle$
<b>1</b>	1.2070	59.724	126.052	190.173	125.316
<b>2</b>	1.9410	69.981	133.740	217.685	140.468
<b>3</b>	2.2520	71.662	143.055	223.776	146.164
<b>4</b>	3.4063	73.733	154.362	222.370	150.155
<b>5</b>	2.3882	75.640	165.574	230.133	157.115
<b>6</b>	5.1947	78.509	173.896	226.310	159.571

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